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The half-lives of some electric quadrupole transitions in the lead region

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With 7 figures in the text

ABSTRACT

Measurements of half-lives of isomeric nuclear states have been carried out on electromagnetically isotope separated samples with a 25-channel time analyzer. The 25-channel analyzer made it possible to investigate short-lived mother nuclei. In the cases below the half-lives of the mother nuclei varied from 1.5 h up to 20 h.

The following half-lives were obtained:

The 1383 kev level of Pb^{202m}: $T_{\frac{1}{2}} = 2.0 \pm 0.15$ mμs
The 148 kev level of Tl^{200}: $T_{\frac{1}{2}} = 7.1 \pm 0.15$ mμs
The 173 kev level of Tl^{198}: $T_{\frac{1}{2}} = 4.5 \pm 1$ mμs

For the following levels an upper limit has been determined:

The 367 kev level of Tl^{199}: $T_{\frac{1}{2}} \leqslant 1.5$ mμs
The 330 kev level of Tl^{201}: $T_{\frac{1}{2}} \leqslant 1.5$ mμs

In all measurements NaI(Tl) scintillation detectors have been used.

Introduction

As is well known, most of the investigations of transition probabilities of electric quadrupole transitions have been undertaken in the region of the isotopic chart where rotational states are expected to be found. In the vicinity of closed shells on the other hand not so many cases are known. It is therefore of special interest to make a further investigation of this region. A search for $E2$ transitions with measureable half-lives in the lead region has been undertaken here.

Most nuclei with new measurable isomeric states have lifetimes of the order of hours. Therefore, in order to obtain low statistical errors and to avoid large decay corrections, it is convenient to use some kind of multi-channel time-analyzer. Such an analyzer gives, in any case, the most reliable results because nothing has to be changed during the measurements and because fluctuations in the counting rates of the detectors do not influence the result, due to the fact that the whole coincidence resolution curve is taken up at the same time.

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In the investigation reported here concerning proton bombarded thallium samples, a 25-channel time-analyzer has been used [1]. Because this type of bombardment gives rise to a sequence of lead isotopes some form of isotope separation was necessary. This was performed in an electromagnetic isotope separator [2]. Since such a separation gives low activity yield, it was convenient to use NaI(Tl) crystals as detectors.

Instrument and method

1. *The coincidence arrangement*

The measurements were carried out using a 25-channel time-analyzer. This arrangement has been described in detail elsewhere [1]. Therefore only a short summary will be given here.

Fig. 1 shows a block-diagram of the apparatus. $\varnothing 1^1/_2'' \times 1''$ NaI(Tl) crystals mounted on RCA 6342 photomultipliers have been used in all measurements.

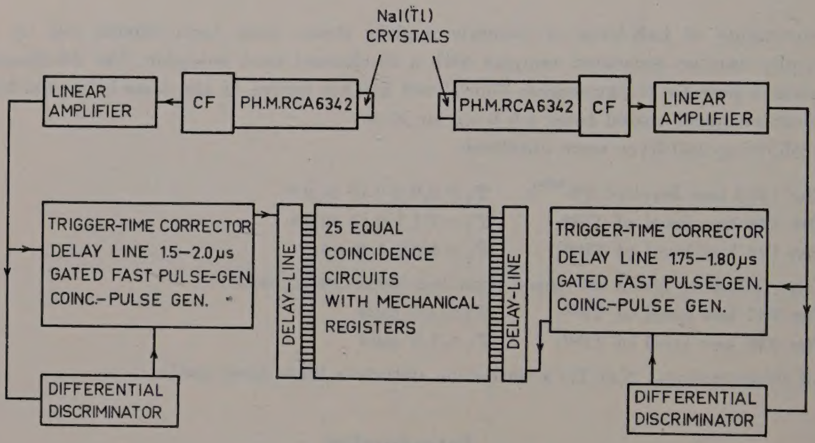

Fig. 1. Block diagram of the 25-channel time analyzer.

The multiplier pulses are fed to an ordinary linear amplifier (Rise-time 0.25 μs). The output pulses from this amplifier are fed to a unit containing a trigger-time corrector, a gated, fast pulse-generator and a coincidence-pulse generator. The amplified multiplier pulses are allowed to pass through the gated fast pulse-generator only if the differential discriminator has delivered a gate pulse. Thus, only pulses selected by the discriminator circuit are triggering the coincidence-pulse generator. The coincidence-pulses are then fed to a unit consisting of 25 equal coincidence circuits. In order to prevent more than one register from being actuated at the same time the register unit is provided with an anticoincidence system between the channels. This gives thus 25 subsequent channels lying side by side with no overlapping or discontinuous jumps between them. The time-separation between the channels is carried out by an exchangeable lumped delay-line.

2. *The coincidence resolution curve*

The coincidence curve obtained with the 25-channel analyzer will be a differentiated time function, while the coincidence curve obtained with usual techniques is an integrated time function. Because it is almost impossible to make the channel widths the same, a special integration method is used [1]. This method transforms the differentiated curve into an integrated one and assigns to the channel numbers certain positions on the time-axis. Fig. 2 demonstrates a typical recording of the half-life of the 481 kev level in Hf^{181}. The lower curve shows the registered number of coincidences in each channel. This curve results from time-spread in the crystals and the photomultipliers, and from the time distribution caused by the measured half-life. The slope of the distribution curve is a measure of the half-life provided that the "half-life" of the slope of a corresponding prompt curve is much less than the actual half-life. As can be seen from the lower curve in Fig. 2 the recorded number of coincidences in a given channel is a function of the channel-width. Thus, a time-distribution curve

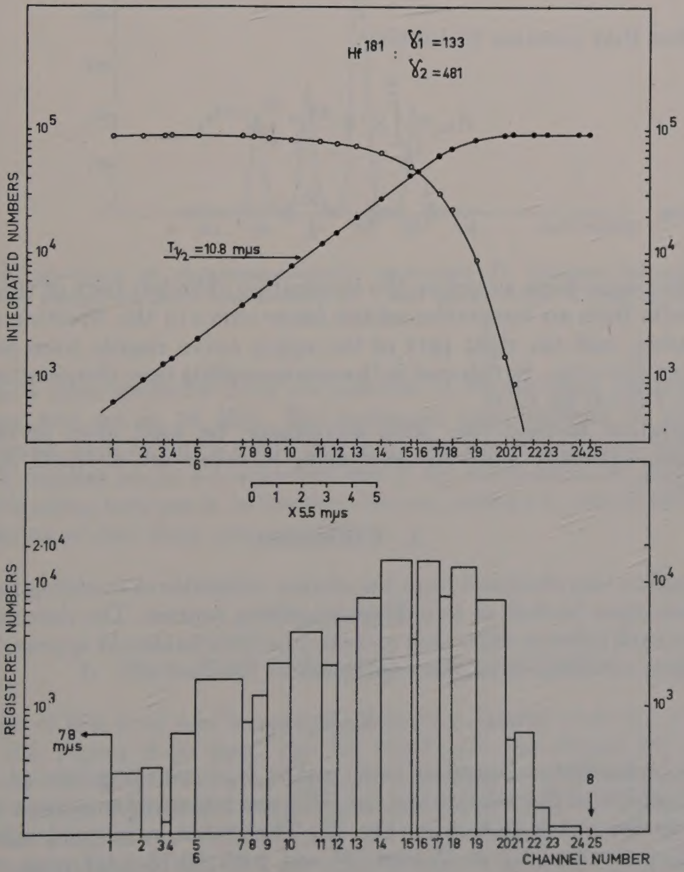

Fig. 2. Coincidence resolution curve obtained for a Hf^{181} sample. The upper curve represents an integration of the lower pile-curve.

cannot directly be drawn; but if the following integration is carried out the channel number will correspond to a certain position on the time-axis, and the integrated number of coincidences will only be a function of this position.

If a_n is the recorded number in the channel n then the integrated number, A_n, will be $A_n = a_n + a_{n+1} \dots a_{25}$ or $A_n = a_n + a_{n-1} \dots a_1$ If the time distribution, $f(t)$, is assumed to be an exponential function, then the number A_n will be

$$A_n = \int_{t_n}^{t_{25}} \varkappa \cdot e^{-\lambda t} \, dt \quad \text{or} \quad A_n = \int_{t_1}^{t_n} \varkappa \cdot e^{-\lambda t} \, dt,$$

depending on the integration direction [$f(t) = \varkappa \cdot e^{-\lambda t}$ for $t > 0$ and $f(t) = 0$ for $t < 0$; $t = 0$ somewhere in the middle of the channel range. $\varkappa$ = constant and λ = decay constant]. The first expression gives the delayed side of the coincidence-curve:

$$A_n = \frac{\varkappa}{\lambda} (e^{-\lambda t_n} - e^{-\lambda t_{25}}).$$

If the number that remains to infinity,

$$A_\infty = \int_{t_{25}}^{\infty} \varkappa \cdot \mathrm{e}^{-\lambda t} = \frac{\varkappa}{\lambda} \mathrm{e}^{-\lambda t_{25}},$$

is added, then

$$A_n + A_\infty = \frac{\varkappa}{\lambda} \mathrm{e}^{-\lambda t_n},$$

which is of the same form as before the integration. The left part of the upper curve in Fig. 2 results from an integration of the lower curve in the direction of decreasing channel number, and the right part of the upper curve results from an integration in the reverse direction. In this case is the corresponding time distribution $f(t) = \varkappa \cdot e^{\lambda t}$ for $t < 0$ and $f(t) = 0$ for $t > 0$.

This integration method can, with advantage, be used even in usual half-life measurements, especially when the measuring intervals are unequal. Such a case is shown in Fig. 7.

3. *Calibration*

The calibration was obtained from the chance-coincidence counting rate appearing when the detectors looked at two different strong sources. The detectors were well shielded from each other in order that no real coincidences should appear. The channel-range was then obtained from the expression:

$$\tau = N_{\mathrm{ch}} / n_1 \cdot n_2,$$

N_{ch} = chance coincidence counting rate, and n_1, n_2 = counting rate at the discriminators. The half-life of the 481 kev level of Hf^{181} was measured as a check of the calibration and gave the value 10.8 mμs (Fig. 2). This value is in good agreement with 10.6 ± 0.3 mμs obtained by de Waard [3] and with 10.75 ± 0.1 mμs reported elsewhere [11]. In order to save time during the measurements Hf^{181} was used as a calibration sample before and after each measurement.

4. *Preparation of spectrometer sources*

All the investigated isotopes have been produced in the Uppsala synchrocyclotron by the reaction Tl(p,xn)Pb. Different bombarding energies were used in order to reach maximum yield for the isotope investigated. After bombardment Pb had been chemically separated from Tl, following a procedure described e.g. in [4]. A few mg of $PbCl_2$ carrier was then added to the carrier-free Pb activity in order to make it possible to use it as charge material in the ion source of the electromagnetic isotope separator.

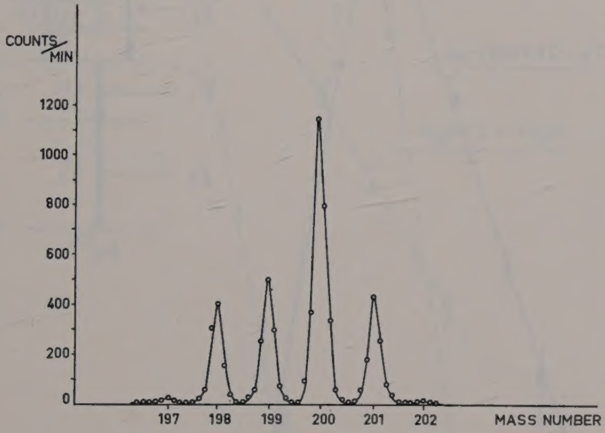

Fig. 3. Activity spectrum of electromagnetically separated Pb isotopes formed in Tl(p,xn) reactions. Proton energy 50 Mev. The activity of 1 mm broad strips is plotted as a function of mass number.

Fig. 3 gives a mass spectrum from a separation of Pb activities obtained when the proton energy was set at 50 Mev. The spectrum was obtained by collecting the isotopes on strips, each having a width of 1 mm. The activity of these was then measured in a GM-counter about 6 h after the end of the bombardment. The contamination by neighbouring isotopes is, in the present case, about 1 %, which can be tolerated in investigations of the kind concerned here.

Measurements

1. *The half-life of the 1383 kev level in Pb^{202m}*

The half-life of this level has been investigated by Åström *et al.* [5], who found no half-life in the region 5–50 mμs, and by Wertheim and Pound [6] who claimed that the half-life should be less than 1 mμs. Our measurement, carried out with electromagnetically isotope-separated samples, gave the value 2.0 ± 0.15 mμs. Fig. 4 shows a coincidence resolution curve obtained in one of the measurements. In this case one channel was set to accept 658 keV γ-rays (γ_1) and the other, 961 kev γ-rays (γ_2). The result of this run is represented by the filled circles. The open circles represent a prompt coincidence curve obtained with a Bi^{207} sample. The 569 kev γ-rays in Bi^{207}

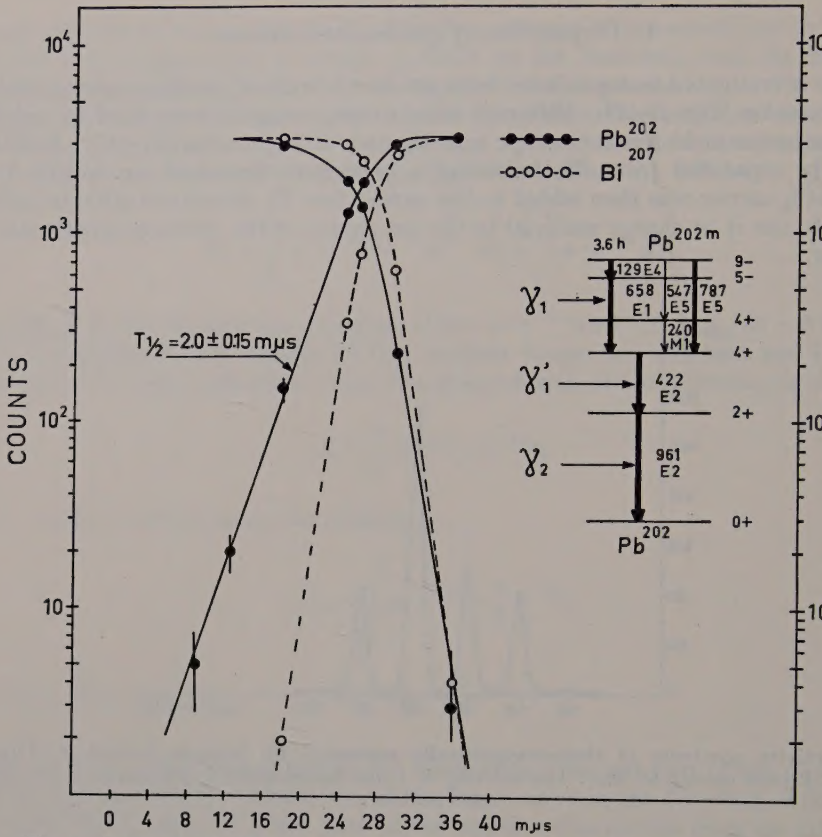

Fig. 4. Delayed coincidence resolution curve for the 1383 kev level in Pb^{202m}.

are shown by Gerholm [7] to have a half-life of 0.09 mμs, which can be said to be prompt compared to 2 mμs. Since the cascade 1064–569 kev in Bi^{207} has almost the same energy as the cascade 658–961 kev in Pb^{202m}, only very small changes in the settings had to be made between the prompt and the delayed runs. Also the source strengths were nearly equal at the beginning of the measurements. Since the Pb^{202m} coincidence curve then decayed with a half-life of about 3.5 hours, no other isotope could have been present in a disturbing amount. Measurements with one channel set to accept 422 kev γ-rays (γ_1) and the other 961 kev γ-rays (γ_2) gave a prompt coincidence curve.

The curve shown in Fig. 4 is drawn after the integration mentioned in the preceding chapter. It is shown elsewhere [8] that the displacement of the intersection point of the two integrated curves with respect to a prompt curve is approximately equal to the mean lifetime of the decay. In the actual case the mean lifetime is of the order of the half-width of the prompt curve. In such a case the displacement of the intersection point is somewhat less than the mean lifetime. In this case the displacement is about $0.9 \cdot \frac{1}{\lambda}$. A calculation of the half-life from this displacement gives 1.95 ± 0.3 mμs which is in good agreement with that obtained from the slope.

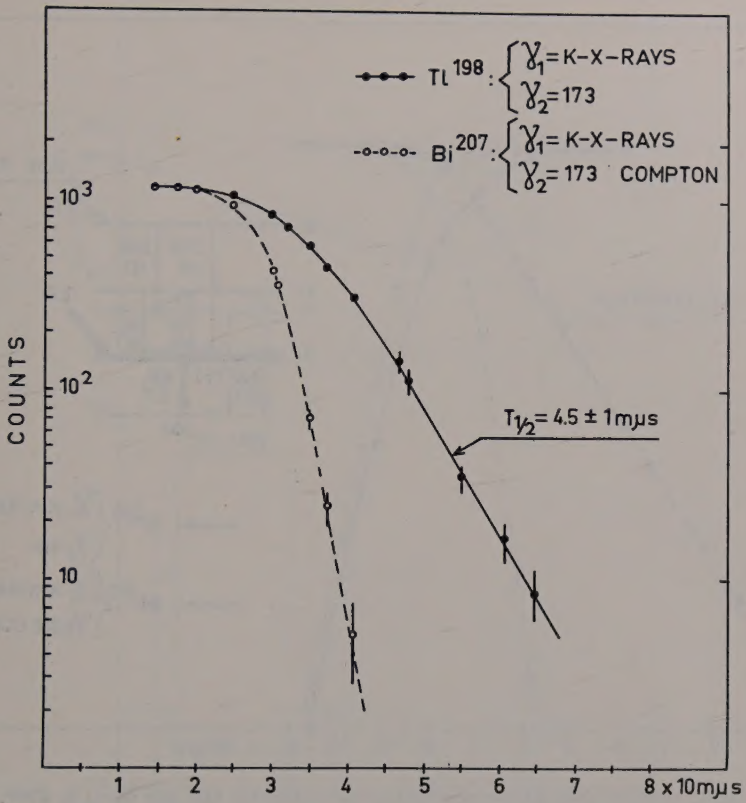

Fig. 5. Delayed coincidence resolution curve obtained for the 173 kev level of Tl^{198}.

2. *The half-life of the 173 kev level of Tl^{198}*

Not very much is known about Pb^{198}, which decays mainly by electron capture to Tl^{198}. Andersson *et al.* [9] and more recently Jung [10] have shown that in this decay there is a strong 173 kev $E2$ transition, which probably is mainly directly fed by electron capture. This should therefore be a case similar to Tl^{200}, which has a first excited state of 148 kev energy and a half-life of 7.1 mμs. Coincidence measurements on an isotope-separated Pb^{198} sample showed that K-X-rays were in strong coincidence with a gamma line of 173 kev. Lifetime measurements of this level resulted in a half-life of 4.5 ± 1 mμs. A curve from one run is shown in Fig. 5. This curve was measured before the 25-channel analyzer was provided with the anticoincidence system between the channels.

3. *The half-life of the 148 kev level in Tl^{200}*

Previous measurements of the half-life of this level gave the value 8 ± 2 mμs [5]. We have remeasured it with the 25-channel analyzer in order to obtain improved accuracy. The result of our measurement was 7.1 ± 0.15 mμs (see Fig. 6).

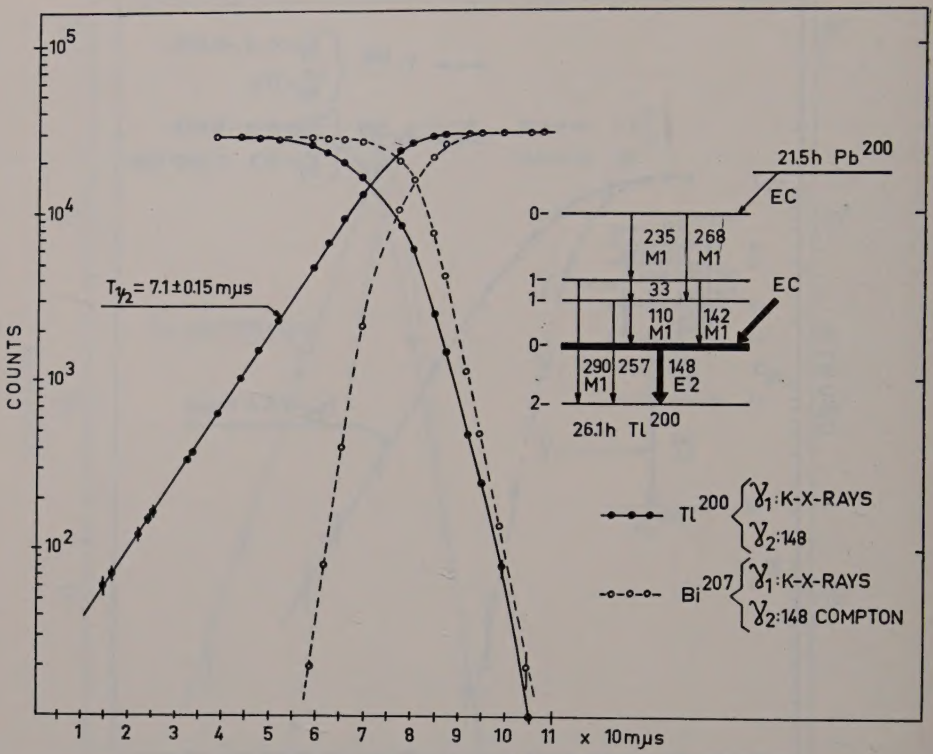

Fig. 6. Delayed coincidence resolution curve for the 148 kev level in Tl^{200}.

Because the 148 kev level is mainly directly fed by electron capture from the ground state of Pb^{200} the investigation was carried out with one detector accepting K-X-rays and the other 148 kev γ-rays. At these low energies the time-spread caused by the crystals is quite large. If it is assumed that the first photo-electron, emitted from the photo-cathode of the photo-multiplier, determines the start-time of an event, then the time distribution will be approximately exponential. The half-life of such a distribution will be about 2 mμs for 70 kev γ-rays and about 1.3 mμs for 150 kev γ-rays. This gives rise to an asymmetrical prompt resolution curve. It is shown elsewhere [11] that this asymmetry does not give any contribution to the actual slope outside the range of the prompt curve if the half-life of this is much larger than the half-life of the corresponding slope of the prompt curve. Furthermore, the slope of the opposite side, actually caused by K-X-rays, has no influence at all on the slope under investigation. The half-life of the 148 kev level can thus be determined with good accuracy from the slope of the delayed curve shown in Fig. 6.

During the measurement the half-life of the mother isotope was followed by measuring the decay of the middle position of the slope of the delayed curve. The result is shown in Fig. 7; the value 20 ± 2 hours was obtained. The delayed curve obviously belongs to the decay of Pb^{200} which has a half-life determined by other methods, of 21.5 hours.

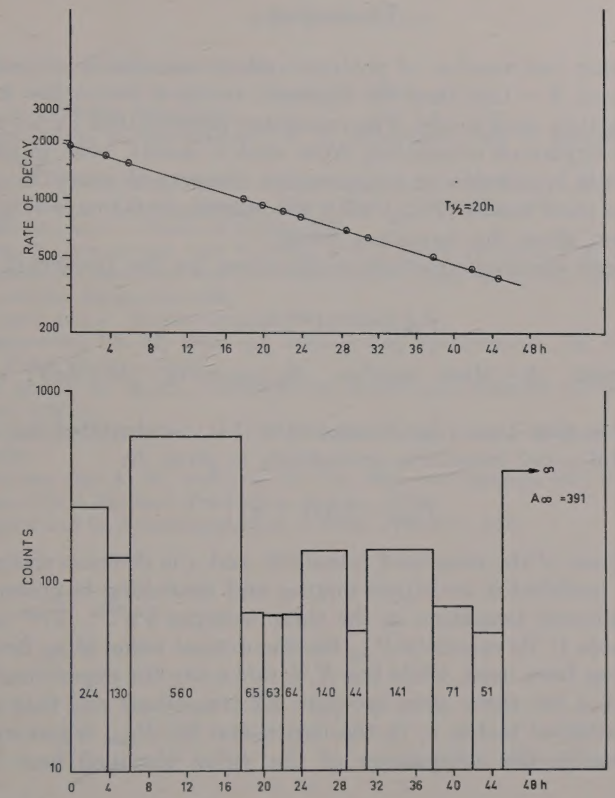

Fig. 7. Rate of decay of the slope of the delayed coincidence curve in Fig. 6. The upper curve is an integration of the lower pile-curve.

4. *The half-life of the 367 kev transition in Tl^{199}*

Pb^{199} decays with a half-life of 1.5 hours to Tl^{199} by electron capture [9]. The first excited state in Tl^{199} is a $d_{3/2}$ state which decays by a mixture of $E2$ and 1-forbidden $M1$ transitions to the $s_{1/2}$ ground state. Delayed coincidence measurements showed that the half-life of this level must be less than 1.5 mμs.

5. *The half-life of the 330 kev level Tl^{201}*

The sequence of isotopes obtained in the isotope separation of the irradiated thallium also contained 9-hour Pb^{201}. This isotope decays with a cascade, similar to that of Pb^{199}, to Tl^{201}. The first excited state has an energy of 330 kev and decays by mixed $E2$ and 1-forbidden $M1$ transitions to the ground state. An upper limit of the half-life of this level was determined to be less than 1.5 mμs.

Discussion

A nuclei, in which the number of protons and of neutrons is close to the magic numbers $Z = 82$ and $N = 126$, may be expected to show properties in accordance with the single particle shell model. The transition probabilities for an $E2$ transition cannot usually be explained completely from such a model, even in the vicinity of a closed shell, but it is suitable to compare the theoretical transition probabilities obtained from the shell model (P_{theo}) with the experimental ones (P_{exp}). The ratio $F = P_{\text{exp}}/P_{\text{theo}}$ then gives the deviation factor.

For $E2$ transitions the single particle model gives for the transition probabilities [12]

$$P_{\text{theo}} = C\,A^{4/3}\,E_\gamma^5\,s, \tag{1}$$

where: C = Constant, A = Mass number, E_γ = γ-energy in MeV, s = statistical factor.

The value of C is that found by Moszkowski [14], recalculated for $r_0 = 1.2$.

The experimental γ-ray transition probability is given by

$$P_{\text{exp}} = 1/\tau(1+\alpha), \tag{2}$$

where τ is the lifetime of the measured transition and α is the conversion coefficient. Eq. (2) has to be modified if multipole mixing and branching is present.

Values of the isomeric transition in the three isotopes Pb^{202m}, Tl^{200} and Tl^{198} are summarized in Table 1. To calculate P_{exp} the theoretical value of α_K from the tables of Sliv *et al.* [13] has been used, while the K/L ratios are the experimental ones. It is further assumed that the three cases are pure $E2$ transitions and that no branching is present. The statistical factor, s, in the expression for P_{theo} is put equal to one in order to make possible the comparison of the value obtained here with that of earlier work.

Table 1

Nucleus	E_γ Mev	$T_{1/2}$ sec	α	P_{exp} sec^{-1}	P_{theo} sec^{-1}	F
Pb^{202m}	0.422 [14]	$2.1 \pm 0.1 \times 10^{-9}$	0.045	0.32×10^9	1.24×10^9	0.26 ± 0.03
Tl^{200}	0.148 [5]	$7.10 \pm 0.15 \times 10^{-9}$	1.23	4.4×10^7	6.23×10^6	7.1 ± 0.4
Tl^{198}	0.173 [9]	$4.5 \pm 1 \times 10^{-9}$	0.65	1.1×10^8	1.38×10^7	8.0 ± 2.4

From the table it is seen that the deviation factor decreases when approching the magic numbers $Z = 82$, $N = 126$, which is just the shell model effect. The values of F are close to the values of the deviation factor for other $E2$ transitions in this region [15, 16].

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